電気材料技術雑誌 第9巻第2号
J.Soc.Elect.Mat.Eng. Vol.9, No.2, 2000

Stimulated Emission and Lasing in π-Conjugated Polymer Films, Microstructures and Opal Photonic Crystals

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Organic materials continue to attract researchers as promising semiconducting media for future electronics. After a recent demonstration[1] of high optical gain and stimulated emission (SE) in films of several π-conjugated polymers, these materials have been intensively studied worldwide as possible gain media for laser applications. Considerable attention has been drawn to lasing in various optical cavities containing luminescent π-conjugated polymers, as well as smaller organic molecules. In our studies of the 2,5-dioctyloxy poly(ρ-phenylene-vinylend)[DOO-PPV] polymer with high photoluminescence (PL) efficiency we demonstrated the occurrence of various SE regimes, such as amplified spontaneous emission (ASE) and lasing. In this contribution we summarize the optical properties of cylindrical, high-Q polymer microcavities such as microring and microdisk laser, and show that light emitting polymer microdiodes with cylindrical geometries are possible candidates for electrically driven plastic lasers. In addition we also report on SE and lasing in opal photonic crystals infiltrated by a variety of polymer and dye solutions. For polymer films we also discuss two unusual SE phenomena, which lead to super-narrow laser-like emission lines without any cavity involved. These phenomena are random lasing[2] and stimulated Raman gain[3].

There are two classes of π-conjugated polymers:luminescent and nonluminescent. The optical properties of the luminescent polymers are very similiar to those of regular organic laser dyes, such as Rhodamines or Coumarins. The primary excitations in these polymers are excitons, which may produce PL with high quantum yield, η, defined as the ratio between the number of emitted photons to the number of absorbed photons. Typically, the radiative decay channel of excitons has to compete with various channels of nonradiative decay; as a result, η is always less than unity. In our studies we have focused on two varieties of luminescent conducting polymers with particularly high PL yields, namely dioctyloxy-PPV (DOO-PPV) with $\eta \approx 0.2$ and poly(1-Phenyl-2-p-n-butylphenylactylene)[PDPA-nBu] with $\eta \approx 0.4$. The Stokes-shifted PL bands, which are broadened by both homogeneous (phonons) and inhomogeneous (disorder) contributions, appear in the spectral range where the ground state absorption is weak. Therefore, a simple exciton model has been adopted to describe the optical transitions responsible for absorption and emission in these polymers. According to this model, the lowest excitations in π-conjugated polymers form a 4-level system, where

transitions 1→2 and 3→4 describe the absorption and emission processes, respectively. The phonon-assisted relaxations 2→3 and 4→1 are very efficient and occur within 100 fsec and 1 psec, respectively. On the other hand, the exciton lifetime, i.e. the decay (mostly nonradiative) time of level 3, in our polymer films is in the range of 100 ps to 1 ns. We conclude that it is possible to achieve inversion between the levels 3 and 4 and thus satisfy the main requirement for a laser medium.

The polymer residual absorption, α_{res}, at the optical frequency between levels 3 and 4 determines the threshold excitation density that is necessary for the population inversion and consequently, lasing. It is preferable, therefore, to have α_{res} as low as possible. However, one of the primary requirements for a polymer laser medium is its high PL yield. It has been noticed that in general, η determined not only by the exciton lifetime, but also by the exciton generation yield. The later may also be less than unity due to the formation of interchain excitations and other nonradiative species. It is essential, therefore, for achieving high η to have both long exciton lifetime and low generation yield of nonemissive excitations. The last fundamental condition influencing the performance of a polymer as a SE medium is the spectral extent of the excitonic photoinduced absorption (PA) in photopumped lasers, and current induced absorption (CIA) of polarons in electrically deiven lasers. The PA or CIA spectra may overlap with the spectrum of the stimulated emission (SE) and thus cancel the total optical gain. It has been shown, however that the PA and SE spectra of singlet excitons in DOO-PPV are well separated, indicating that this polymer can be a good candidate for photopumped laser media.

In our studies of laser action in π-conjugated polymers we have observed ASE in thin films, micro-cavities and polymers infiltrated in opal photonic crystals[2]. True lasing has been achieved in micro-cavities such as micro-rings and micro-disks, but also in opal photonic crystals via the mechanism of distributed feedback aided by phase flips. We also observed random lasing in disordered films or in polymers infiltrated in micro-crystalline opals. In addition we also report on switching that occurs in very thin DOO-PPV films and in polyfluorene films[4] after thermal cycling. The switching occurs between the narrow line emissions associated with 0-0 and 0-1 transitions.

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